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**LETTER TO THE EDITOR**

# The oxygen-isotope effect on the in-plane penetration depth in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ as revealed by muon-spin rotation

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**Abstract**
The oxygen-isotope ($^{16}O/^{18}O$) effect (OIE) on the in-plane penetration depth $\lambda_{ab}(0)$ in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ was studied by means of muon-spin rotation. A pronounced OIE on $\lambda_{ab}^{-2}(0)$ was observed with a relative isotope shift of $\Delta\lambda_{ab}^{-2}/\lambda_{ab}^{-2} = -5(2)\%$ for $x = 0.3$ and $-9(2)\%$ for $x = 0.4$. The OIE exponents of $T_c$ and of $\lambda_{ab}^{-2}(0)$ exhibit a relation that appears to be generic for cuprate superconductors.

The pairing mechanism of high-temperature superconductivity (HTSC) remains elusive in spite of the fact that many models have been proposed since its discovery. A fundamental question is whether lattice effects are relevant for the occurrence of HTSC. In order to clarify this point a large number of isotope effect studies on the superconducting transition temperature $T_c$ have been performed since 1987 [1–10]. It was found that the oxygen-isotope effect (OIE) exponent $\alpha_O = -\mathrm{d}\ln T_c/\mathrm{d}\ln M_O$ shows a generic trend for various cuprate families [1, 4–6, 8–10]: in the underdoped region $\alpha_O$ is large, even exceeding the conventional BCS value $\alpha = 0.5$, and becomes small in the optimally doped and overdoped regimes. A similar trend was also observed for the copper-isotope exponent $\alpha_{Cu}$ [6, 8, 10].

There is increasing evidence that a strong electron–phonon coupling is present in cuprate superconductors, which may lead to the formation of polarons (bare charge carriers accompanied by local lattice distortions) [11, 12]. One way to test this hypothesis is to demonstrate that the effective mass of the supercarriers $m^*$ depends on the mass $M$ of the lattice atoms. This is in contrast to the case for conventional BCS superconductors, where $m^*$ is independent of $M$. For cuprate superconductors (the clean limit) the in-plane penetration

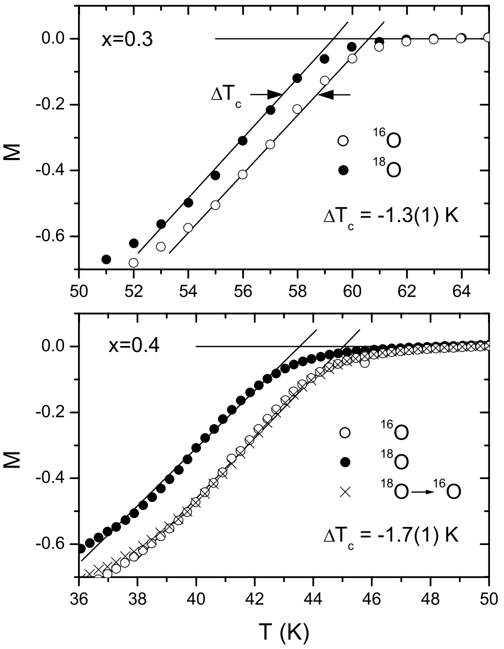

**Figure 1.** A section near $T_c$ of the low-field (1 mT, field-cooled) magnetization curves (normalized to the value at 10 K) for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4).

depth $\lambda_{ab}$ is given by $\lambda_{ab}^{-2}(0) \propto n_s/m^*_{ab}$, where $n_s$ is the superconducting charge carrier density, and $m^*_{ab}$ is the in-plane effective mass of the superconducting charge carriers. This implies that the OIE on $\lambda_{ab}$ is due to a shift in $n_s$ and/or $m^*_{ab}$:

$$\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = \Delta n_s/n_s - \Delta m^*_{ab}/m^*_{ab}. \qquad (1)$$

Therefore a possible mass dependence of $m^*_{ab}$ can be tested by investigating the isotope effect on $\lambda_{ab}$, provided that the contribution of $n_s$ to the total isotope shift is known.

Previous OIE studies of the penetration depth in $YBa_2Cu_3O_{7-\delta}$ [13], $La_{2-x}Sr_xCuO_4$ [9, 14, 15], and $Bi_{1.6}Pb_{0.4}Sr_2Ca_2Cu_3O_{10+\delta}$ [16] indeed showed a pronounced oxygen-mass dependence on the supercarrier mass. However, in all these experiments the penetration depth was determined indirectly from the onset of magnetization [13, 16], from the Meissner fraction [9, 14], and from magnetic torque measurements [15]. The muon-spin rotation ($\mu$SR) technique is a direct and accurate method for determining the penetration depth in type II superconductors. In this letter, we report $\mu$SR measurements of the in-plane penetration depth $\lambda_{ab}$ in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4) with two different oxygen isotopes ($^{16}O$ and $^{18}O$). A large OIE on $\lambda_{ab}$ was observed which mainly arises from the oxygen-mass dependence of $m^*_{ab}$.

Polycrystalline samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4) were prepared by standard solid-state reaction [17]. Oxygen-isotope exchange was performed during heating the samples in $^{18}O_2$ gas. In order to ensure that the thermal histories of the substituted ($^{18}O$) and unsubstituted ($^{16}O$) samples were the same, in all cases two experiments (on $^{16}O_2$ and $^{18}O_2$) were performed simultaneously. The exchange and back-exchange processes were carried out at 600 °C for 25 h, and then the samples were slowly cooled (20 °C h$^{-1}$) in order to oxidize them completely. The $^{18}O$ content in the samples, as determined from a change of the sample weight after the isotope exchange, was found to be 78(2)% for both samples. The total oxygen content

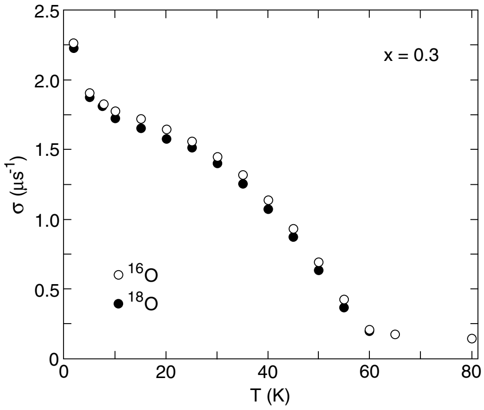

**Figure 2.** The temperature dependence of the $\mu$SR depolarization rate $\sigma$ of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ for $x = 0.3$, measured in a field of 200 mT (field cooled). The error bars are smaller than the size of the data points.

**Table 1.** The oxygen content $y = 7 - \delta$ of the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4) samples determined by high-accuracy volumetric analysis.

|  | $^{16}O$ | $^{18}O$ | $^{18}O \rightarrow {}^{16}O$ (back-exchange) |
|---|---|---|---|
| $x$ | $y$ | $y$ | $y$ |
| 0.3 | 6.942(2) | 6.941(2) |  |
| 0.4 | 6.929(3) | 6.934(3) | 6.933(3) |

of the samples was determined using high-accuracy volumetric analysis [17]. The $^{16}O$ and $^{18}O$ samples for $x = 0.3$ and 0.4 were prepared simultaneously under the same conditions and had within experimental error the same oxygen contents (see table 1). To examine the quality of the samples low-field (1 mT, field cooled), SQUID magnetization measurements were performed (see figure 1). For both concentrations the $T_c$-onset for the $^{16}O$ samples was higher than for $^{18}O$ with nearly the same transition width. An oxygen back-exchange of the $^{18}O$ sample ($x = 0.4$) resulted within error in almost the same magnetization curve as for the $^{16}O$ sample, confirming that the back-exchange is almost complete. The results on the OIE on $T_c$ are summarized in table 2. Taking into account an isotope exchange of 78%, we find $\alpha_O = 0.22(4)$ for $x = 0.3$ and $\alpha_O = 0.37(5)$ for $x = 0.4$, in agreement with previous results [5, 18].

The transverse-field $\mu$SR experiments were performed at the Paul Scherrer Institute (PSI), Switzerland, using the $\pi$M3 $\mu$SR facility. The samples consisted of sintered pellets (12 mm in diameter, 3 mm thick) which were mounted on a $Fe_2O_3$ sample holder in order to reduce the background. The polycrystalline $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples were cooled from far above $T_c$ in a magnetic field of 200 mT. For a powder sample the magnetic penetration depth $\lambda$ can be extracted from the muon-spin depolarization rate $\sigma(T) \propto 1/\lambda^2(T)$, which probes the second moment $\langle \Delta B^2 \rangle^{1/2}$ of the probability distribution of the local magnetic field function $p(B)$ in the mixed state [19]. For highly anisotropic layered superconductors (such as the cuprate superconductors), $\lambda$ is mainly determined by the in-plane penetration depth $\lambda_{ab}$ [19]: $\sigma(T) \propto 1/\lambda_{ab}^2(T) \propto n_s/m_{ab}^*$.

The depolarization rate $\sigma$ was extracted from the $\mu$SR time spectra using a Gaussian relaxation function $R(t) = \exp[-\sigma^2 t^2/2]$. Figure 2 shows the temperature dependence of the

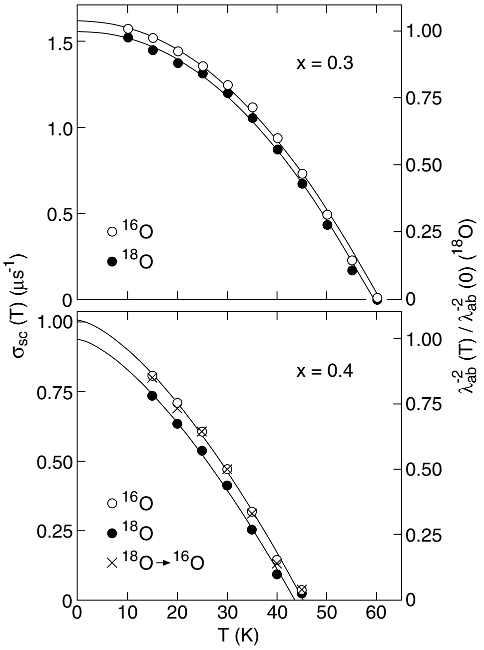

**Figure 3.** The temperature dependence of the depolarization rate $\sigma_{sc}$ in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ for $x = 0.3$ and 0.4 (200 mT, field cooled). On the right-hand axis the normalized in-plane penetration depth $\lambda_{ab}^{-2}(T)/\lambda_{ab}^{-2}(0)(^{18}O)$ is plotted for comparison with [15]. The solid curves correspond to fits to the power law $\sigma_{sc}(T)/\sigma_{sc}(0) = 1 - (T/T_c)^n$. The error bars are smaller than the size of the data points.

measured $\sigma$ for the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with $x = 0.3$. Similar results were obtained for the samples with $x = 0.4$. It is evident that the values of $\sigma$ for $^{18}O$ are systematically lower than those for $^{16}O$. As expected for a type II superconductor in the mixed state, $\sigma$ continuously increases below $T_c$ with decreasing temperature [19]. The sharp increase of $\sigma$ below $\simeq$10 K is due to antiferromagnetic ordering of the Cu(2) moments [20]. However, zero-field $\mu$SR experiments indicate for both $x = 0.3$ and 0.4 samples no presence of magnetism above 10 K. Above $T_c$ a small temperature-independent depolarization rate $\sigma_{nm} \simeq 0.15\ \mu s^{-1}$ is seen, arising from the nuclear magnetic moments [19]. Therefore, the total $\sigma$ is determined by three contributions: a superconducting ($\sigma_{sc}$), an antiferromagnetic ($\sigma_{afm}$), and a small nuclear magnetic dipole ($\sigma_{nm}$) contribution. Because the contribution $\sigma_{afm}$ is only present at low temperatures, data points below 10 K were excluded from the analysis. The superconducting contribution $\sigma_{sc}$ was then determined by subtracting $\sigma_{nm}$ measured above $T_c$ from $\sigma$. In figure 3 we show the temperature dependence of $\sigma_{sc}$ for the $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with $x = 0.3$ and 0.4. It is evident that for both concentrations a remarkable oxygen-isotope shift of $T_c$ is present, as well as that of $\sigma_{sc}$.

The data in figure 3 were fitted to the power law $\sigma_{sc}(T)/\sigma_{sc}(0) = 1 - (T/T_c)^n$ [19] with $\sigma_{sc}(0)$ and $n$ as free parameters, and $T_c$ taken from the magnetization measurements (see table 2). The values of $\sigma_{sc}(0)$ obtained from the fits are listed in table 2 and are in agreement with previous results [20]. The exponent $n$ was found to be $n = 2.0(1)$ for $x = 0.3$ and $n = 1.5(1)$ for $x = 0.4$, which is typical for underdoped YBCO [19]. Moreover, $n$ is within error the same for $^{16}O$ and $^{18}O$. This implies that $\sigma_{sc}$ has nearly the same temperature dependence for the two isotopes (see figure 3). In order to prove that the observed OIE on

**Table 2.** Summary of the OIE results for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ extracted from the experimental data (see the text for an explanation).

| | $^{16}$O | | $^{18}$O | | | | |
|---|---|---|---|---|---|---|---|
| $x$ | $T_c$ (K) | $\sigma_{sc}(0)$ ($\mu\mathrm{s}^{-1}$) | $T_c$ (K) | $\sigma_{sc}(0)$ ($\mu\mathrm{s}^{-1}$) | $\alpha_{\mathrm{O}}$ | $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0)$ (%) | $\beta_{\mathrm{O}}$ |
| 0.3 | 60.6(1) | 1.63(2) | 59.3(1) | 1.57(2) | 0.22(4) | −5(2) | 0.38(12) |
| 0.4 | 45.3(1) | 1.01(2) | 43.6(1) | 0.94(2) | 0.37(5) | −9(2) | 0.71(14) |
| 0.4 | 45.1(1)[a] | 1.01(4)[a] | | | | | |

[a] Results for the back-exchange ($^{18}\mathrm{O} \to {}^{16}\mathrm{O}$) sample.

$\lambda_{ab}(0)$ is intrinsic, the $^{18}$O sample with $x = 0.4$ was back-exchanged ($^{18}\mathrm{O} \to {}^{16}\mathrm{O}$). As seen in figure 3, the data points for this sample (cross symbols) do indeed coincide with those for the $^{16}$O sample. From the values of $\sigma_{sc}(0)$ listed in table 2, the relative isotope shift of the in-plane penetration depth $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = [\sigma_{sc}^{18\mathrm{O}}(0) - \sigma_{sc}^{16\mathrm{O}}(0)]/\sigma_{sc}^{16\mathrm{O}}(0)$ was determined. Taking into account an isotope exchange of 78%, one finds $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0) = -5(2)$ and $-9(2)$% for $x = 0.3$ and 0.4, respectively (table 2). For the OIE exponent $\beta_{\mathrm{O}} = -\mathrm{d}\ln\lambda_{ab}^{-2}(0)/\mathrm{d}\ln M_{\mathrm{O}}$, one readily obtains $\beta_{\mathrm{O}} = 0.38(12)$ for $x = 0.3$ and $\beta_{\mathrm{O}} = 0.71(14)$ for $x = 0.4$ (table 2). This means that for underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ the OIE on $\lambda_{ab}^{-2}$, like that on $T_c$, increases with increasing Pr doping $x$ (decreasing $T_c$). This finding is in excellent agreement with the recent magnetic torque measurements on underdoped $La_{2-x}Sr_xCuO_4$ [15].

According to equation (1) the observed $\Delta\lambda_{ab}^{-2}(0)/\lambda_{ab}^{-2}(0)$ is due to a shift of $n_s$ and/or $m_{ab}^*$. For $La_{2-x}Sr_xCuO_4$, several independent experiments [9, 14, 15] have shown that the change of $n_s$ during the exchange procedure is negligibly small. In the present work we provide further evidence: (i) The fully oxygenated $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples ($\delta \simeq 0$) were all prepared under identical conditions, either in a $^{16}O_2$ or a $^{18}O_2$ atmosphere [17], and the Pr content $x$ did not change. It is very unlikely that $n_s$ changes significantly upon $^{18}$O substitution, and after the back-exchange ($^{18}\mathrm{O} \to {}^{16}\mathrm{O}$) the same results are obtained (see figures 1, 3 and table 2). (ii) According to a model [21] that describes the suppression of $T_c$ in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$, the number of supercarriers decreases linearly with increasing $x$ in the range of $0.05 < x < 0.5$, and consequently $\Delta n_s/n_s = -\Delta x/x$. Moreover, for $0.1 < x < 0.5$ the transition temperature $T_c$ decreases linearly with $x$, with $\Delta T_c/\Delta x \simeq -150$ K/Pr atom [5]. Combining these two relations one obtains: $\Delta T_c \simeq -150x\,\Delta n_s/n_s$. Assuming that the observed OIE on $\lambda_{ab}^{-2}$ is due only to a change of $n_s$ ($\Delta m_{ab}^*/m_{ab}^* \simeq 0$), one can estimate the corresponding shift of $T_c$. For $x = 0.3$ and 0.4 one finds $\Delta T_c \simeq -1.8(4)$ and $-4.2(6)$ K, respectively. The experimental values, however, are much lower (see figure 1): $\Delta T_c = -1.3(1)$ K ($x = 0.3$) and $\Delta T_c = -1.7(1)$ K ($x = 0.4$). We thus conclude that any change in $n_s$ during the exchange procedure must be small, and that the change of $\lambda_{ab}$ is mainly due to the OIE on the in-plane effective mass $m_{ab}^*$ with $\Delta m_{ab}^*/m_{ab}^* \simeq 5(2)$ and 9(2)% for $x = 0.3$ and 0.4, respectively. This implies that the effective supercarrier mass $m_{ab}^*$ in this cuprate superconductor depends on the mass of oxygen atoms, which is not expected for a conventional phonon-mediated BCS superconductor. To our knowledge there are only two theoretical models of HTSC which predict an OIE on the effective carrier mass ($m^*$), namely, a bipolaronic model of Alexandrov and Mott [11] and a model of nonadiabatic superconductivity proposed by Grimaldi *et al* [22].

In figure 4 the exponent $\beta_{\mathrm{O}}$ versus the exponent $\alpha_{\mathrm{O}}$ for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ is plotted. For comparison we also included the recent magnetic torque results for underdoped $La_{2-x}Sr_xCuO_4$ [15]. It is evident that these exponents are linearly correlated: $\beta_{\mathrm{O}} = A\alpha_{\mathrm{O}} + B$. A best fit yields $A = 1.8(4)$ and $B = -0.01(12)$, so $\beta_{\mathrm{O}} \simeq A\alpha_{\mathrm{O}}$. This empirical relation appears to be generic for cuprate superconductors. One can understand this behaviour in

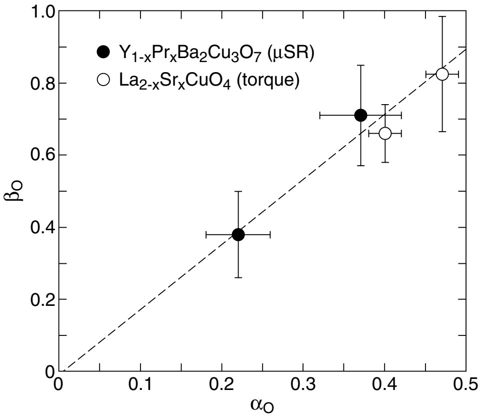

**Figure 4.** A plot of the OIE exponent $\beta_O$ versus the OIE exponent $\alpha_O$ for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3$ and 0.4) and $La_{2-x}Sr_xCuO_4$ ($x = 0.080$ and 0.086) from [15]. The dashed line represents a best fit to the empirical relation $\beta_O = A\alpha_O + B$.

terms of an empirical relation between $T_c$ and the $\mu$SR depolarization rate $\sigma_{sc}(0)$ [23, 24]. It was shown [24] that for most families of cuprate superconductors the simple parabolic relation $\bar{T}_c = 2\bar{\sigma}(1 - \bar{\sigma}/2)$ describes the experimental data rather well (here $\bar{T}_c = T_c/T_c^m$, $\bar{\sigma} = \sigma_{sc}(0)/\sigma_{sc}^m(0)$, and $T_c^m$ and $\sigma_{sc}^m(0)$ are the transition temperature and the depolarization rate of the optimally doped system). Using this parabolic ansatz, one readily obtains the linear relation between $\beta_O$ and $\alpha_O$: $\beta_O/\alpha_O = 1 + 1/2[(1 - (1 - \bar{T}_c)^{1/2})/(1 - \bar{T}_c)^{1/2}]$.

In the heavily underdoped regime ($\bar{T}_c \to 0$), $\beta_O/\alpha_O \to 1$. For the underdoped samples shown in figure 4 the reduced critical temperature $\bar{T}_c$ is in the range 0.5–0.7, yielding $\beta_O/\alpha_O = 1.2$–$1.4$, in agreement with $A = 1.8(4)$ obtained from the experimental data. Recently, it was pointed out [25] that the unusual doping dependence of the OIE on $T_c$ and on $\lambda_{ab}^{-2}(0)$ naturally follows from the doping-driven 3D–2D crossover and the 2D quantum superconductor-to-insulator transition in the underdoped limit. It is predicted that in the underdoped regime $\beta_O/\alpha_O \to 1$, which is consistent with the parabolic ansatz.

In summary, we performed $\mu$SR measurements of the in-plane penetration depth $\lambda_{ab}$ in underdoped $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0.3, 0.4$) for samples with two different oxygen isotopes ($^{16}$O and $^{18}$O). A pronounced OIE on both the transition temperature $T_c$ and $\lambda_{ab}^{-2}(0)$ was observed, which increases with decreasing $T_c$. The isotope shift of $\lambda_{ab}^{-2}(0)$ is attributed to a shift in the in-plane effective mass $m_{ab}^*$. For $x = 0.3$ and 0.4 we find $\Delta m_{ab}^*/m_{ab}^* = 5(2)$ and 9(2)%, respectively. Furthermore, an empirical relation between the OIE exponents $\beta_O$ and $\alpha_O$ was found that appears to be generic for various classes of cuprate superconductors. The OIE on $m_{ab}^*$ implies that the superconducting carriers have polaronic character, and that lattice effects play an essential role in the occurrence of HTSC.

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